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AIRBORNE AND GROUND-BASED MEASUREMENTS OF ATMOSPHERIC PARTICLES FROM CLUSTERS TO SUB-MICROMETER SIZES

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Academic dissertation

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Katri Elisabet Leino
University of Helsinki, 2019

Abstract

Atmospheric aerosol particles in different sizes have different impacts on climate and humans. Particularly in high concentration they may cause various negative health effects for people due to e.g. toxicity. Larger particles impact on the climate by absorbing or scattering the solar radiation and participating in cloud formation in the atmosphere. Some of atmospheric gases nucleate forming molecular clusters and grow in size to secondary aerosols. The phenomenon is called new particle formation (NPF) and it is contributing up to 50 % of the total aerosol number budget globally.

In this thesis has focused on exploring NPF at different layers of atmosphere using a small aircraft as a platform for flying instrumentation and comparing with the data measured at ground level, SMEAR II, Hyytiälä. Based on the airborne and ground-based observations also the spread of wildfire emissions from Eastern Europe to Finland was studied.

In case of studying NPF measurement flights over the SMEAR II (up to 3500 meters) were conducted during several campaigns between 2009 and 2017. We found out that NPF frequently occurs within the whole boundary layer (BL) and that the vegetation could be a key contributor in producing aerosol precursor gases in boreal environment. However, some signs were also found that NPF could occur separately in the residual layer or lower free troposphere.

Emission sources produce different gases and particles into the atmosphere near the ground. Emissions can be divided to natural and anthropogenic. Example of a natural source is uncontrolled biomass burning in the forests and peat bogs, which is frequently observed around the world during warm and dry seasons. Air pollution from biomass burning is thought to have a significant influence on the chemical composition of the atmosphere and Earth's climate system. In this study we investigated long-range transported air pollution in Finland originating from wildland fires in Eastern Europe in 2006 and 2010. The fire emissions were detected as increased concentrations of different gases and total number of particles at three SMEAR stations in Finland. The smoke plumes where detected also at higher altitudes during flight measurement campaign in summer of 2010.

This study improves the understanding of the first steps of atmospheric NPF inside the BL and elucidates the impacts of air pollution originating from forest fires on air quality and climate after long-transport.

Contents

List of publications

This thesis consists of an introductory review, followed by 4 research articles. The articles are cited according to their roman numerals.

I. Schobesberger, S., Väänänen, R., **Leino, K.,** Virkkula, A., Backman, J., Pohja, T., Siivola, E., Franchin, A., Mikkilä, J., Paramonov, M., Aalto, P. P., Krejci, R., Petäjä, T. and Kulmala, M.: Airborne measurements over the boreal forest of southern Finland during new particle formation events in 2009 and 2010. Boreal Environ. Res., 18, 145–163, 2013.

II. **Leino, K.,** Lampilahti, J., Poutanen, P., Väänänen, R., Manninen, A., Buenrostro Mazon, S., Dada, L., Franck, A., Wimmer, D., Aalto, P. P., Ahonen, L., Kangasluoma, J., Keronen, P., Korhonen, F., Laakso, H., Matilainen, T., Siivola, E., Manninen, H. E., Lehtipalo, K., Kerminen, V.-M., Petäjä, T. and Kulmala, M.: Vertical profiles of sub-3 nm particles over boreal forest. Atmos. Chem. Phys., 19, 4127–4138, 2019.

III. **Leino, K.,** Nieminen, T., Manninen, H. E., Petäjä, T., Kerminen, V.-M. and Kulmala, M.: Intermediate ions as a strong indicator of new particle formation bursts in a boreal forest. Boreal Environ. Res., 21, 3–4, 274–286, 2016.

IV. **Leino, K.,** Riuttanen, L., Nieminen, T., Dal Maso, M., Väänänen, R., Pohja, T., Keronen, P., Järvi, L., Aalto, P. P., Virkkula, A., Kerminen, V.-M., Petäjä, T. and Kulmala, M.: Biomass-burning smoke episodes in Finland from eastern European wildfires. Boreal Environ. Res., 19 (suppl. B), 275–292, 2014.

1 Introduction

According to the definition, a mixture of liquid or solid aerosol particles that float in a carrier gas, such as air, is called an aerosol (Hinds, 1999). Thus, the air in the Earth's atmosphere is also an aerosol. It consists of different gases with small amount of particulate matter, aerosol particles (Barry and Chorley, 2010). The biggest parts of dry air by volume are nitrogen, (N_2, 78.08 %), oxygen, (O_2, 20.98 %) and argon, (Ar, 0.93 %). The remaining part consists carbon dioxide, (CO_2, 0.04 %) and other gases. All atmospheric gases, except of nitrogen and oxygen are called trace gases due to their small concentration.

Different gases and particulate matter are emitted into the atmosphere near the Earth's surface. The emissions can be called natural or anthropogenic, depending on the emission sources. Wildland fires produce smoke (Ward and Hardy, 1991; Alves et al., 2010), volcanic eruptions ash (Gudmundsson et al., 2012), Saharan desert and other dry areas dust (Perez et al., 2008; Schepanski et al., 2012), oceans sea salt (Grythe et al., 2014) and many other processes emit gases and aerosols into the atmosphere through natural processes. Biomass burning (Li et al., 2010) and other combustion processes (Myung and Park, 2012), transport (Viana et al., 2014) and agricultural activity are examples of human-initiated sources of gases and particulate matter into the air.

The gases which are contributing to the Earth's radiation budget by absorbing thermal radiation and causing an increase in air temperature (greenhouse effect) are called greenhouse gases. Without any greenhouse effect, the average temperature on our planet would be over 30 degrees lower. The most important greenhouse gases on the Earth are water vapor (H_2O) and carbon dioxide (CO_2), but also methane (CH_4), nitrous oxide (N_2O), and ozone (O_3) have a warming effect (Rotmans, 1990). The lifetime of different greenhouse gases varies, which gives them different weight on their contribution to the global climate change. The lifetime of water vapor is only few days, methane 9–15 years, nitrous oxide 120 years and carbon dioxide even hundreds of years (Archer et al., 2009). The amount of atmospheric carbon dioxide has increased year by year and it is estimated that the effects will be irreversible for 1000 years although the emissions would decrease (Solomon et al., 2009).

The climate change is observed to speed up melting of snow and ice cover which is harmful especially for Arctic areas (Comiso et al., 2008). It causes rising of sea level and induces stronger storms and other extreme weather phenomena (Jentsch and Beierkuhnlein, 2008). The climate change generates more inequality on living conditions for the societies, like food and water supply and threatens the ecosystems and biodiversity particularly in the tropics (Corlett, 2012). The main reason for the large

increase in the greenhouse gas concentrations into the atmosphere is human-initiated emissions (IPCC, 2018).

The aerosol particles can be divided into two groups according to their formation mechanisms. Primary aerosols are emitted into the atmosphere directly as particles in processes like biomass burning and wind-driven dust resuspension, whereas secondary aerosols are formed from gas phase precursors in the atmosphere via gas-to-particle conversion (Kroll and Seinfeld, 2008; Kulmala and Kerminen, 2008; Hallquist et al., 2009; Kulmala et al., 2013; Kulmala et al., 2014).

While the greenhouse gases have a warming effect on the climate, the aerosol particles, on average, cool the Earth's climate total (IPCC, 2018), except for black carbon (BC) particles that strongly absorb radiation thus to warm the climate. The effects can be divided to direct and indirect effect. Current IPCC terminology divides these to aerosol radiation interactions (ARI, direct) and aerosol cloud interactions (ACI, indirect). Atmospheric aerosols scatter and absorb the coming sunlight and backscattered longwave radiation directly and large enough aerosol particles may act as cloud condensation nuclei (CCN) affecting the climate indirectly. Depending on the number and size of cloud droplets, the age and the composition of them differ and thus also the effect on radiation balance on the Earth. Particles play a key role in cloud formation and thus water cycle on our planet.

In addition to the climate effect, increased amount of aerosol particles deteriorates air quality and visibility and affects negatively on people's health. The air quality impacts are seen especially in polluted urban environments (Chan and Yao, 2008; Kan et al., 2012) and for instance during intensive wildfires (van der Werf et al., 2010). The aerosol particles (from ultrafine sizes (< 0.1 μm) up to coarse particles (even 10 μm)) in the air that we breathe have been observed to be reason for many allergies, asthma and cardiovascular and respiratory diseases (Nel, 2005; Franck et al., 2011; Kim et al., 2015; WHO, 2016).

The size range of atmospheric aerosol particles vary from newly-formed 1 nm molecular clusters up to 100 μm coarse particles (Fig. 1.1). The total number concentration of atmospheric aerosol particles can vary from some tens in very clean environments, like in Antarctica to 10^8 particles in cubic centimeter in polluted megacities (Koponen et al., 2003; Kulmala et al., 2005, 2016; Pey et al., 2008; Manninen et al., 2010; Kumar et al., 2014). In addition to the total number concentration, size distribution is a simple way to describe the size and number, area or volume concentration of the aerosol population.

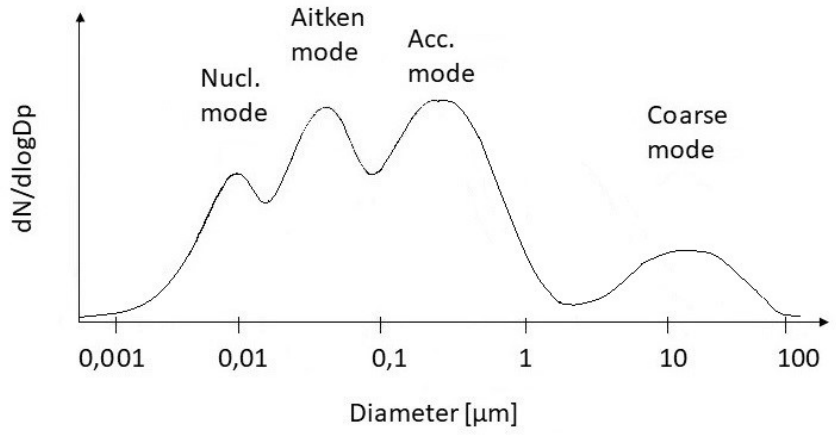

Figure 1.1. A schematic representation of atmospheric aerosol number size distribution as a function of particle diameter. In addition, a cluster mode occurs in size range below 3 nm.

In contrast to the greenhouse gases, the atmospheric lifetime of the aerosol particles including black carbon (BC) is quite short. The largest coarse mode particles feel the impact of gravitational force, so they deposit onto surfaces quickly. The lifetime of newly formed 1–2 nm clusters (cluster mode) and the smallest nucleation mode particles is also relatively short due to efficient removal by coagulating or deposition due their high diffusivity. These particles grow larger by multicomponent condensation (e.g. Riipinen et al., 2012). Growing in size, the nucleation mode particles reach the Aitken mode. The accumulation mode particles have the longest lifetime and therefore they can be transported long distances. Their lifetime in the atmosphere is few weeks.

One of the important sources of aerosol number concentration in the Earth's atmosphere is New Particle Formation (NPF) (Mäkelä et al., 1997; Kavouras et al., 1998; Weber et al., 1999; Birmili et al., 2003; Kulmala et al., 2004; Holmes, 2007; Kulmala et al., 2007; Kulmala et al., 2013; Rose et al., 2015; Bianchi et al., 2016; Kerminen et al., 2018, Chu et al., 2019). NPF is regional rather than a local phenomenon. Newly-formed nanoscale particles are formed in the atmosphere by clustering of precursor vapors and grow in size by multi-component condensation (Kulmala et al., 1998; Zhang et al., 2012; Riipinen et al., 2012; Ehn et al., 2014). It was estimated that even half of global cloud condensation nuclei (CCN) is originally formed via secondary aerosol formation (Spracklen et al., 2008; Merikanto et al., 2009; Luo and Yu, 2011).

In addition to formation and growth mechanisms, nucleation, condensation and coagulation, the aerosol particles are influenced by several removal processes. The aerosol particles can be removed

from the atmosphere by dry or wet deposition. The wet deposition is a removal process of aerosol particles via activation to cloud droplets and scavenging due to raindrops, snow and cloud droplets. In dry deposition, the particles collide and impact, fall down due to gravity or diffuse caused by turbulence or Brownian motion. The deposition is the most effective removal process for the smallest and largest atmospheric aerosols.

Condensation sink (CS) is a useful parameter in analyzing formation and growth of atmospheric newly formed particles (Dal Maso et al., 2002). It describes the rate at which the condensable vapors are lost into the aerosol phase. From the vapor point of view, the inverse of CS is descriptive of vapor lifetime before it is scavenged by the aerosol population. Coagulation sink, in turn, is the size dependent parameter that describes the ability of aerosol population to remove particles of a certain size due to collisions. The coagulation is the most effective for particles with a large size difference so it reduces effectively the number of particles and grows the mean size of the particles as the small particles move by Brownian motion and coagulate with the larger, slowly moving particles which have large surface areas.

In this study I investigate the formation of secondary aerosol particles and their growth in boreal environment and explore their vertical, horizontal and temporal variability. I also explore the effects of long range-transported air pollution originated from forest fires (Fig. 1.2) to the concentrations aloft and at the ground level. The NPF was studied over boreal forest in Southern Finland (**Papers I–III**) and long range-transported fire emissions at three different measurement stations over Finland during 2006 and 2010 large-scale wild fires in Eastern Europe (**Paper IV**). In both cases a manned aircraft was used as a platform for the measurement instrumentation.

The aims of this study are:

- to investigate the temporal and spatial extent of NPF at boreal forest site in Southern Finland (**Papers I and II**)
- to develop a more objective identification method for the NPF events (**Paper III**)
- to investigate and characterize long-transported air pollution in the lower atmosphere originated from forest fires and biomass burning (**Paper IV**)

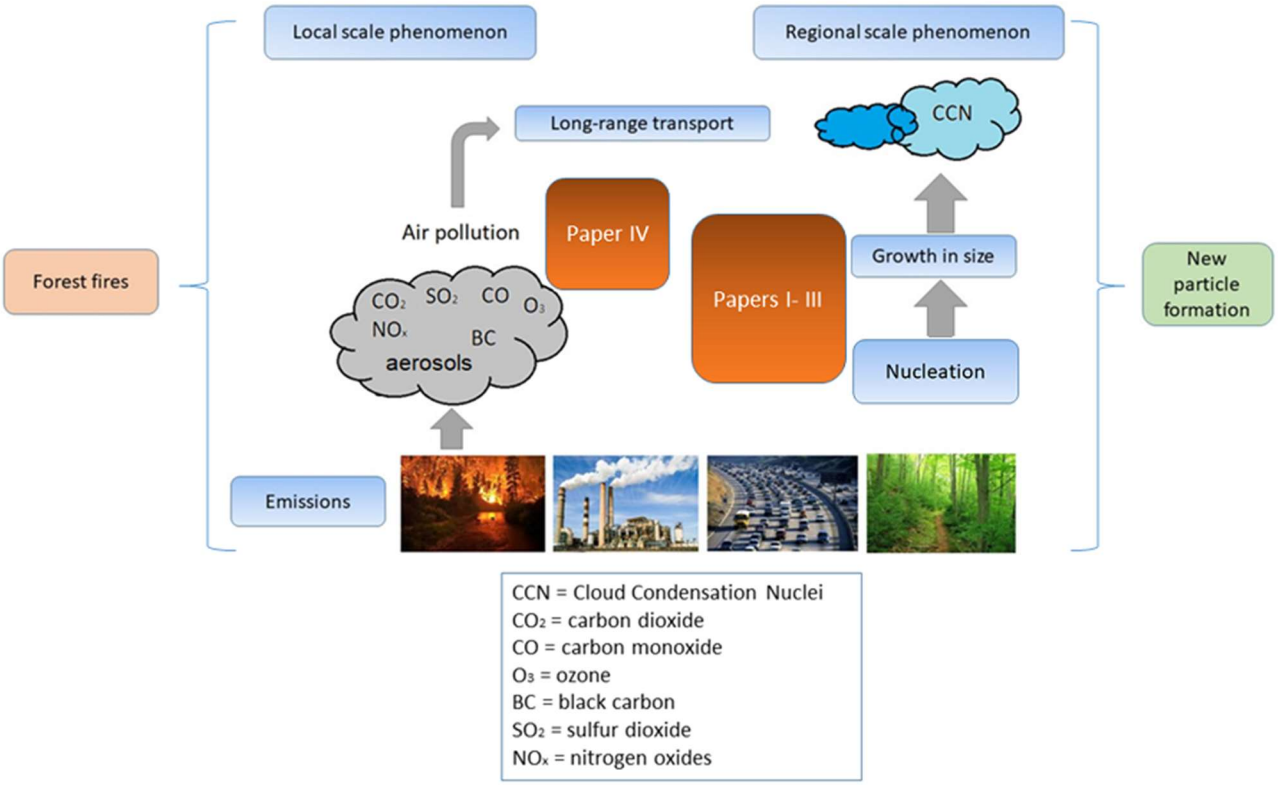

Figure 1.2. The themes and connections pertinent to this thesis.

2 Atmospheric boundary layer – structure and evolution

Atmospheric turbulence is an irregular flow of motion of air mass that is generated by thermally driven vertical currents (convection) or wind shear. When the air masses move near an irregular terrain or some obstacle, like mountain or building, the motion is disturbed resulting in varying wind speed and direction forming turbulence. This is called mechanical turbulence. Thermal turbulence is induced when the Sun heats the surface causing convective thermals to rise. These thermals displace colder air that starts to mix down. As a result, different sizes of turbulent eddies are formed. The thermally driven turbulence produces also convective clouds, like cumulus and cumulonimbus. Turbulence can be also formed in upper altitudes, where the wind speeds are typically higher because of lower impact of surface friction. Chaotic changes in fluid or gas flow velocity and direction and thus turbulence occur in many size ranges from the molecular scale turbulence to mesoscale roll vortices (Stull, 2012).

Atmospheric boundary layer (ABL) or planetary boundary layer (PBL) over the planetary surface is the closest air surrounding the Earth. The BL structure develops with diurnal cycle represented in Fig. 2.1 and atmospheric turbulence has a significant role in the developing process.

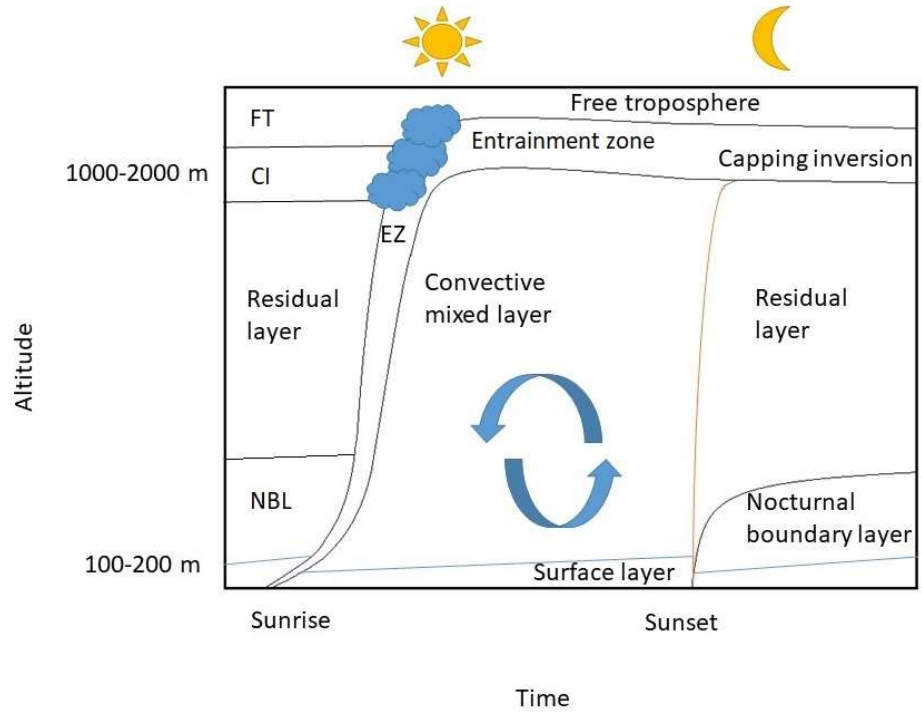

Figure 2.1. A schematic representation of the development of the boundary layer structure during a course of a day according to Stull (2012).

According to the definition, PBL is the air layer which has been directly in connection with the Earth's surface during the past day or it reacts to the changes in surface forcing in one hour or less through exchange of momentum, heat and mass (Stull, 2012). The lowest ~10 % part of PBL near the surface is called surface layer. In the morning the rising sun starts to warm the Earth's surface. The warm surface causes heating in the air above it, which is lifted upward since it is lighter. Cooler and denser air, in turn, descends to displace it. This causes turbulent mixing that stretches higher and higher leading to the growth of a new developing BL. This layer is also called convective mixing layer, ML. The upper boundary of the PBL is rising up within the day reaching the maximum height typically around mid-afternoon.

In the evening the heat driven turbulence ceases and the PBL transforms into a residual layer (RL) after termination of solar heating. At the same time the stable nocturnal boundary layer (NBL) starts to shape. The RL, seen in night time and early next morning, includes the aerosol and gas concentrations from the previous day's BL (Stull, 2012).

The space above the BL and RL is free troposphere (FT). In addition, between BL and FT is an entrainment zone (EZ) that acts as "a port" between turbulent air and more stable FT air. In the entrainment zone, thermal convection transports turbulent BL air upward furthering the rise of the mixed-layer top and at the same time, whereas drier less turbulent air from the FT can, in turn, penetrate down. Between the layers is often a capping inversion, where air temperature increases with height.

ML is typically thermal or surface friction induced turbulent layer, but turbulence is observed also for instance in entrainment zone and outflows of the clouds. Turbulence has been observed to have a strong contribution to atmospheric NPF, enhancing it by creating favorable conditions like fluctuations in temperature and humidity, hence local supersaturation of precursor gases and also diluting the background aerosol concentration, which are essential for NPF.

2.1 Estimation of boundary layer height

In order to investigate the vertical extent of NPF, we need to know the height of BL. Estimating the boundary layer height (BLH) is a complex process and there are many methods and strategies for estimation and verification of the BLH. As we know, turbulence transports the heat, particles, gases, including water vapor. Many measured quantities are suitable to give preliminary estimation for the BLH, but because of the complexity of atmospheric conditions some constraints and challenges occur. The most reliable estimation is derived using different measuring systems and methods together.

Inside the well-mixed BL, quantities like potential temperature (describes the temperature that the air would have, if it would be transferred adiabatically to the reference air pressure) and water vapor mixing ratio (the ratio of the mass of water vapor to the mass of dry air) are vertically quite constant because of strong convective turbulent mixing (Stull, 2012). In an ideal case there is a sharp decrease in vertical profile of relative humidity (RH) or/and concurrent a sharp increase in potential temperature in the transitional region between the ML and RL or FT. The estimation of BLH in this study (**Papers I, II and IV**) is based on measurements on board a research aircraft, but the estimation can be done also from frequent radiosonde profiles (Wang and Wang, 2014), which provide BLH height in a synoptic scale.

Fig 2.2 shows vertical measurement profiles of particle concentration, H_2O mixing ratio, potential temperature and relative humidity (RH) during a Cessna measurement flight at 8:15–9:00 a.m. on 13th August 2015. RH describes how much water vapor is in the air in relation to the highest absolute value that the air may contain at a certain temperature governed by saturation vapor pressure. As we see in Fig. 2.2a, the water evaporates from the surface and it is lifted up inside the ML. At the altitude of 700 meters, the ratio decreases quickly. The vertical profile of CO_2 behaves in a reverse manner (Fig. 6 and 7 in **Paper II**). Inside the BL, the amount of CO_2 is typically lower than above it, due to uptake of CO_2 by the vegetation in forested areas, like Hyytiälä.

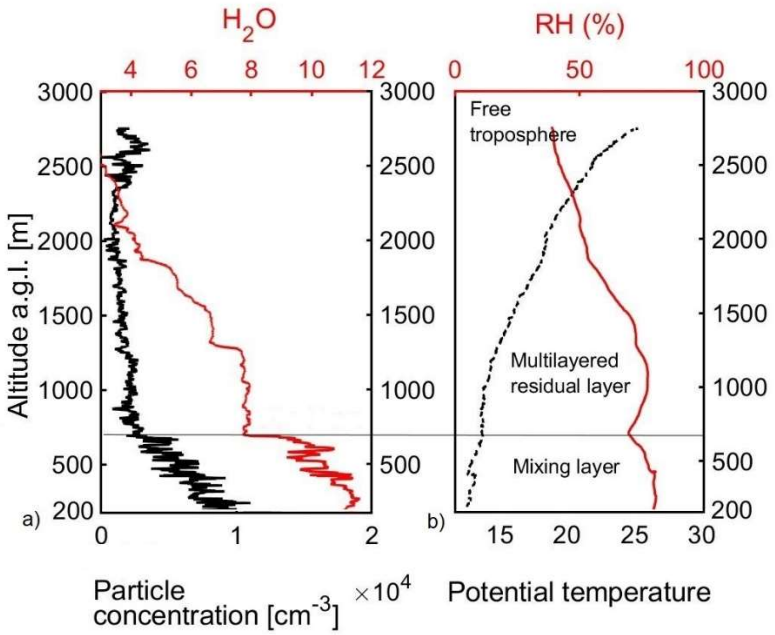

Figure 2.2. Higher concentration of water vapor and aerosol particle number concentration are often seen inside the mixing boundary layer, like in 13.8.2015 in the morning in Hyytiälä. The water vapor mixes up from the ground level within the evolving BL. In addition, residual layers from the previous day BL are frequently seen above the convective ML.

The BLH can be evaluated also from LIDAR (LIght Detection And Ranging) backscatter measurements conducted on the ground level (Hennemuth and Lammert, 2006; Manninen et al., 2016). The LIDAR instrumentation produces pulsed laser light signals, which backscatter from the targets in the air. The intensity and travel time of the backscattered light pulses give information about the targets, like aerosol particles in the air. The LIDAR method is useful for determining BLH.

3 Experimental setup

3.1 Ground stations

The ambient aerosol and gas measurements on the ground level presented in this thesis were conducted at three SMEAR (Station for Measuring Ecosystem-Atmosphere Relations, Hari et al., 2016) around Finland (SMEAR I in Värriö (Hari et al., 1994), SMEAR II in Hyytiälä (Hari and Kulmala, 2005) and SMEAR III in Helsinki (Järvi et al., 2009)). The data from all the three stations is presented in **Paper IV,** whereas data from SMEAR II is analyzed in **Papers I–III**.

SMEAR I is the oldest of the SMEAR stations in Finland (Hari et al., 1994). It is established in 1991 in Värriö (69°46′N, 29°35′E, 390 m a.s.l), in Finnish Lapland. The station is a remote subarctic background station located near Russian border. A 15-m-high measurement tower is located on the top of a hill. From the SMEAR I in this thesis we analyzed aerosol particle number and volume size distributions, condensation sink (CS) and concentrations of CO, O_3, NO_x and SO_2 in order to address the aerosol and gas-phase signatures pertinent to long-transported biomass burning emissions from intensive forest fires in Russia in 2006 and 2010 (**Paper IV**).

SMEAR II is the second oldest of stations (Hari and Kulmala, 2005). It was instituted in 1995 in coniferous forest ambience in Southern Finland (61°51′N, 24°17′E, 181 m a.s.l) about 50 km northeast from the nearest big city, Tampere. The surrounding stand consists mainly of Scots pine with some Norway spruce and deciduous trees. The top of the tree canopy is in around 20 meters. The surroundings of SMEAR II include mainly forest, but also some lakes, peatlands and some agricultural activity and households. The nearby emission sources are vegetation and agriculture, but also other emissions from anthropogenic activity, like industry and traffic and long-range transported pollution from Europe and for instance wildfires are occasionally observed when prevailing wind is therefrom.

The comprehensive instrumentation is located in measurement cottages, containers, on 18- and 35-m-high towers and on 128-m-high mast. The station is globally known to produce high-quality, long-term continuous and comprehensive datasets on atmosphere-ecosystem relations (Hari and Kulmala, 2005). The data from SMEAR II was widely used in this thesis. The total particle size distributions were used in **Papers I, II and IV** and ion size distributions was analyzed in the study of intermediate size range ion concentrations in **Paper III**. Of trace gases, we analyzed CO, CO_2, O_3, NO, NO_x, H_2O and SO_2 in case of biomass burning emission study (**Paper IV**). Also CS and black carbon concentrations were supporting the analysis of the character of the biomass burning smoke. The

calculated sensible heat flux (SHF) was used in **Paper II** as it is indicative for the BL development indicating the strength of turbulent mixing and homogeneity inside the BL.

SMEAR III (60°10´N, 24°57´E, 26 m a.s.l) is a relatively new urban background station located in close vicinity of busy Lahti highway and University of Helsinki Kumpula Campus, four kilometers from the center of Helsinki. The station has been operational since the year of 2004 and it includes a measurement container and a 31-m-high measurement tower (Järvi et al., 2009). The analyzed data from this station in this thesis includes particle number and volume size distributions, CS and the concentrations of NO, NO_x, O_3, SO_2 and CO (**Paper IV**).

3.2 Airborne platform

Airborne measurements in this study were conducted using a single-engine, Cessna FR172F small aircraft (**Papers I, II and IV**). All flights were operated from and to Tampere-Pirkkala airport. The flight time of the plane carrying instrumentation and two people is roughly 3 hours. The flights were flown with an optimal airspeed of 125 km h^{-1}. The vertical measurement profiles from ~100 meters up to 4000 meters were carried out in the surrounding area (< 30 km) of the SMEAR II to get the best comparability between the airborne and on-ground measurements in **Papers I** and **II**. In **Paper I** the studied area was larger (~ 100 km from SMEAR II). The measurement flights in **Paper IV** exploring the spatial extent of biomass burning plumes were conducted in Eastern and Central Finland.

In the Cessna, the back seats of the airplane were replaced by an instrumentation rack. The rack contained a varying set of instrumentation depending on the purpose of the campaign. In 2009 campaign (**Paper I**) total particle number concentrations were measured with three condensation particle counters (CPC battery, Kulmala et al., 2007), including two TSI 3772 CPCs with the cut-off sizes of 6 and 10 nm and one TSI 3776 ultrafine-CPC with cut-off size of 3 nm (Mordas et al., 2008). The time resolution of each CPC was 1s. Starting from 2010 (**Papers II and IV**), TSI 3772 CPCs were replaced by scanning mobility particle sizer (SMPS, Wang and Flagan, 1990). The size range of this number size distribution measurement system was 10–400 nm with 2 min time resolution. In the **Paper II** we measured the total particle concentration with 1.5 nm cut-off size with a new-generation particle counter, Particle Size Magnifier (PSM) (Vanhanen et al., 2011).

The instrumentation included also a triple wavelength (467, 530 and 660 nm) particle/soot absorption photometer (PSAP, Radiance Research) and a nephelometer using a wavelength of 545 nm (Radiance Research Model 903, **Papers I and IV**), H_2O- and CO_2- gas analyzer (Licor LI-840), Global

Positioning system (GPS) receiver, devices for basic meteorological variables, like temperature, pressure, relative humidity (RH), photosynthetically active radiation (PAR) (**Papers I and IV**), and wind speed and 3D-direction (**Paper II**).

The main sample flow for the instruments was collected through 3.3–4.3 m long steel inlet line attached on the right wing of the plane (**Papers I and IV**). The sampling was performed through a scaled-down solid diffuser inlet designed by the University of Hawaii (McNaughton et al., 2007). Starting from 2013 an additional sample line extending 50 cm distance from the fuselage of the airplane (Fig. 3.1) (**Paper II**) was utilized. The main flow inside the sample lines was kept at 47 lpm. Every instrument took the sample from the central line of the main flow minimizing the losses.

Figure 3.1. The inlet line for sample air used in **Paper II** came out from the left back window of the Cessna measurement airplane.

3.3 Instrumentation

3.3.1 Condensation particle counters

For counting the number concentration of sample aerosol particles larger than the minimum detection size of the instrument condensation particle counters (CPC) are largely used. The first CPCs were developed over one hundred years ago (McMurry, 2000). The CPC detects the particles optically, which means that they have to be first grown into a size large enough by vapor condensation. As a working fluid the instruments use water or alcohols, like butanol or isopropanol. Inside the CPC the supersaturated vapor condenses onto the sampled particles. The method to create the supersaturation can be in adiabatic expansion, mixing or diffusion in a laminar flow. The latest mentioned is widely used in commercial CPCs. Its advantages are continuous sampling, relatively small losses and exact temperature control. In the laminar flow CPCs, the aerosol sample is mixed with vaporized working fluid, like n-butanol (**Papers I, II and IV**) in a warmed saturator. After that the mixture is cooled in a condenser and the supersaturated vapor condenses onto the particles. The grown particles are optically counted in a detector. In this study we used three different models of butanol based laminar flow CPC: TSI 3772 (**Papers I and IV**), TSI 3776 (**Papers I, II and IV**) and TSI 3010 (**Paper II**).

Recently a new generation condensation particle counter – Particle Size Magnifier (PSM) (Vanhanen et al., 2011) was developed. The instrument is based on turbulent mixing of heated diethylene glycol (DEG) saturated flow and the colder sample flow for achieving supersaturation. The advantage of the instrument is the ability to measure particles down to ~1 nm size, which is crucial in investigation of the first steps of NPF. This is achieved using DEG as a working fluid, because it has a lower supersaturation vapor pressure and higher surface tension than for example butanol (Iida et al., 2009). However, the PSM is not able to grow the particles large enough for the optical detection. Therefore, a booster CPC needs to be used downstream of the PSM (Vanhanen et al., 2011). Overall, the instrument is also versatile. It can be used in scanning mode or in fixed mode to measure total particle concentration with a tuned cut-off size, depending on the mixing ratio or temperature difference between the saturated flow and sample flow. Scanning mode results the size distribution typically from ~1 nm to ~2–3 nm (Lehtipalo et al., 2014). The PSM system we used in this study (**Paper II**) consists of the PSM (Airmodus, A10) and CPC (TSI 3010) as a counter for the particles.

3.3.2 Differential and scanning mobility particle sizers

Differential Mobility Particle Sizer (DMPS, Aalto et al., 2001) and Scanning Mobility Particle Sizer (SMPS, Wang and Flagan, 1990) are spectrometers, which are widely used to determine aerosol number size distribution of the aerosol population. In this study an airborne SMPS scanned the size distribution in the range from 10 to 400 nm. DMPS, in turn, was measuring on ground level at SMEAR II in the size range of 3–1000 nm.

The principle of both instruments is the same. The aerosol particles are classified according to their electrical mobility equivalent size and their concentration is determined. Both spectrometers include a drier, a bipolar charger (a radioactive source), a Differential Mobility Analyzer (DMA) and a Condensation Particle Counter (CPC). Sometimes a twin-DMPS system is used, like at SMEAR II. The twin-system enables larger measurement size range as a whole, because the whole size range is divided in to two parts which are measured separately with their own DMA and the detector (more details are presented in Aalto et al., 2001). With the bipolar charger the aerosol population is brought to a well-defined bipolar charging state (Wiedensohler, 1988). The DMA consists of inner and outer electrodes and there is an electric field between the electrodes determined by the voltage difference. In this thesis cylindrical DMAs were used (Winklmayr et al., 1991). The charged aerosol particles in aerosol flow feel a force of electric field. Depending on the flows, magnitude of electric field and electrical mobility of particles, they drift toward the inner electrode, wherefrom the particles with exact electrical mobility are led to outgoing sample flow. Adjusting the voltages, the particles with selected electrical mobility can be selected to the sample flow. The electrical mobility depends on the particle charge and size. Small particles have a higher mobility than bigger ones when both have same charge. The number of the particles is counted by CPC.

The main difference between DMPS and SMPS is the measurement protocol how the DMA classifying voltage is changed. The SMPS measures the diameter range changing the voltages smoothly in a continuous manner, whereas the DMPS uses a stepwise variation of voltage. Getting aerosol size distribution from the measured data demands the use of inversion methods. In addition to twin-DMPS system, the other possibility to measure size range in two parts is using two-flow cycles, like in SMPS measurements of this study (Hakala, BSc thesis, 2010). Geometry and flowrates of the DMA have an effect on the size range of the system. We used the aerosol and sheath air flow rates of 1 liters per minute (lpm) and 5 lpm for the size range of 10–40 nm and 4 lpm and 20 lpm for the size range of 40–400 nm, respectively (**Papers I, II and IV**).

3.3.3 Neutral cluster and air ion spectrometer

Neutral cluster and Air Ion Spectrometer (NAIS, Airel Ltd., Estonia) is a very useful instrument for measuring naturally charged ions in the size range of 0.8–42 nm (Mirme et al., 2013; Manninen et al., 2016). It can measure also total particle distribution of aerosol population with a controlled charging of sample in the size range of 2.25–42 nm (Asmi et al., 2009). The larger uncertainties in case of NAIS measurements are caused by a unipolar charging of the aerosol population instead of bipolar charging in case of SMPS and DMPS systems. According to the calibration measurements, the size information given by the NAIS is observed to be very accurate (Wagner et al., 2016). The different data inversion algorithms, used with NAIS measurements showed that the number concentrations determined by the NAIS were typically underestimated in the smaller sizes (0.98–5 nm), whereas the number concentrations at diameters 19.6 nm and larger were overestimated (Wagner et al., 2016).

The NAIS classifies charged particles and ions according to their electrical mobility. The instrument consists two multichannel parallel differential mobility analyzers (DMA) enabling simultaneous measurement of negatively and positively charged particles. The outer electrode of the DMA is divided into 21 insulated collectors. The charged particles and ions drift onto the collector governed by their electrical mobility and produce a current signal. The electrical mobility depends on the particle charge and size. Small particles have a higher mobility than bigger ones when the both have same charge. The mobility range of instrument is 3.16–0.0013 $cm^2\ V^{-1}\ s^{-1}$ corresponding to an electrical mobility equivalent diameter range of 0.8–42 nm (Millikan-Fuchs equivalent diameter; Mäkelä et al., 1996).

The total flowrate of NAIS is 60 or 54 lpm, depending on the model and it is divided for both of columns, 30 or 27 lpm per polarity. Sheath flow rates are 60 lpm in each DMA. The chargers are unipolar corona needles. In a typical measurement setup, the NAIS measures alternately total particles, ions and background noise. The cycle takes approximately 2–3 minutes to scan a whole size scale with all measurement modes.

The particle and ion size distributions were measured at SMEAR II with two NAIS and DMPS systems. Fig. 3.2 represents the corresponding size distribution measured at ground level in Hyytiälä and on-board the Cessna. The airborne aerosol number size distribution differed from the ground observations as the altitude varied during the flight and free tropospheric (FT) air was measured in the middle of both flights. The air mass characteristics in the FT is not connected with the ground level conditions, whereas the air inside the BL was well-mixed within the whole BL. Thus, the aerosol

number size distribution data inside the BL corresponded well with the ground observations, illustrated by the same kind of size distribution at both locations.

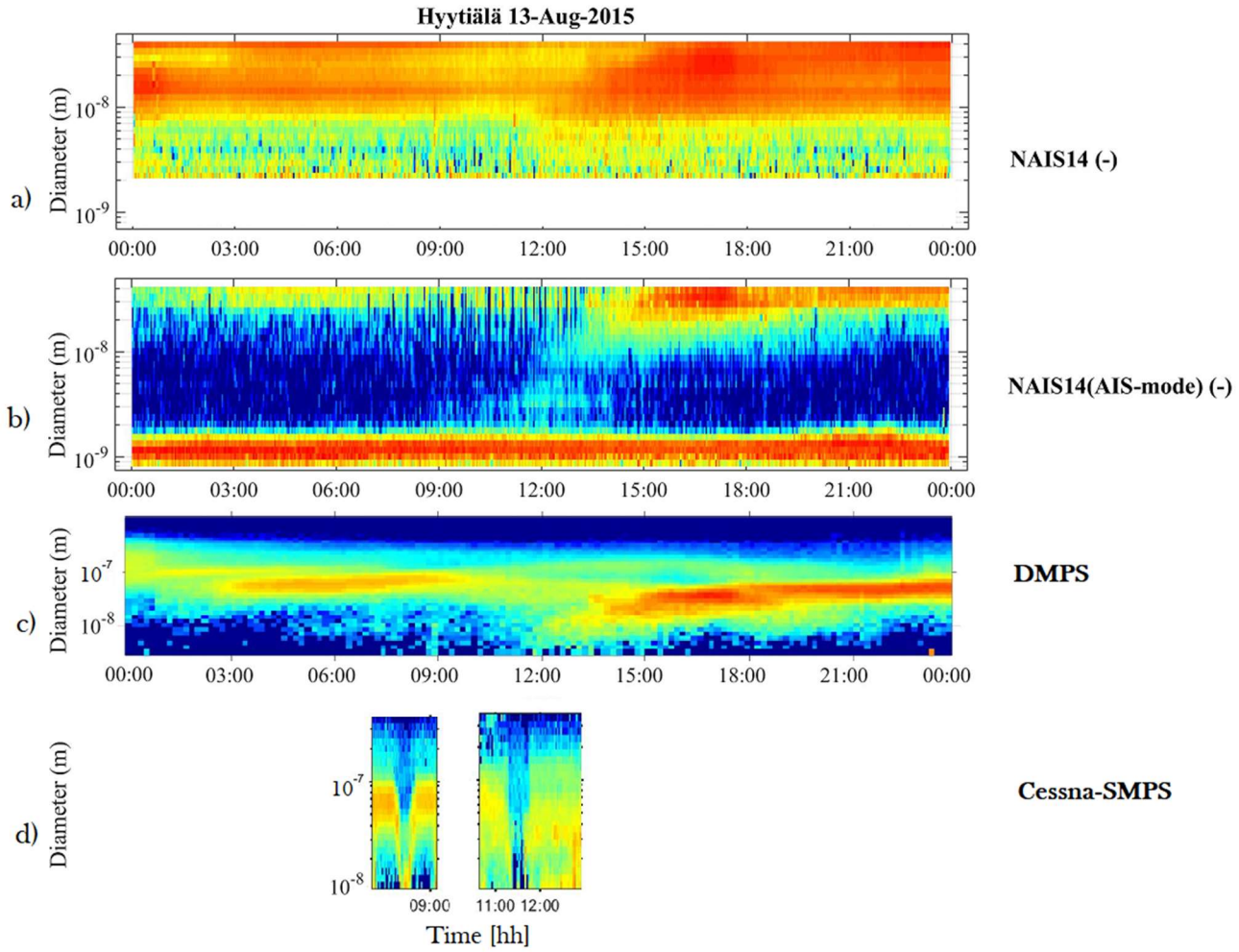

Figure 3.2. These contour plots represent NPF event day 13th August 2015 around SMEAR II. The size distributions at first three plots (a-c) were measured at ground level in Hyytiälä and d was measured on-board Cessna research aircraft over Hyytiälä area during two research flights. The figures a, c and d show total number size distribution and figure b depict naturally charged particles number size distribution. The SMPS-data was collected while the measurement altitude varied. The start of the NPF process is detected both at the ground level and onboard the Cessna before noon.

4 Atmospheric new particle formation

In the formation of new atmospheric aerosol particles, suitable precursor vapors form molecular clusters, which can stabilize and further grow in size by multi-component condensation (Zhang et al., 2012; Kulmala et al., 2014). The phenomenon has been observed to occur in various environments around the world (Covert et al., 1992; Weber et al., 1996, 1997, 1999; Kulmala, 2004; Manninen et al., 2010; Kerminen et al., 2018; Chu et al., 2019). Numerous field measurements have confirmed that NPF events occur in relatively clean environments (Dal Maso et al., 2005; Asmi et al., 2010; Kyrö et al., 2013; Nieminen et al., 2014; Dada et al., 2017), and recently they have been observed also in polluted megacities (Xiao et al., 2015; Yu et al., 2017; Zhang et al., 2017; Yao et al., 2018; Chu et al., 2019).

During last decades several field, laboratory and modelling studies have been conducted to investigate the NPF process, formation and growth mechanisms together with participating gaseous compounds. Sulfuric acid (oxidation product of SO_2) is often associated with the formation and growth of new atmospheric aerosol particles (Weber et al., 1996; Sihto et al., 2006; Petäjä et al., 2009; Sipilä et al., 2010; Yue et al., 2010; Kulmala et al., 2013). However, it is recognized that it cannot alone explain the formation and growth, but other species, like cluster stabilizing compounds; ammonia (Berndt et al., 2010; Kirkby et al., 2011) and amines (Kurtén et al., 2008; Paasonen et al., 2012; Almeida et al., 2013) and oxidized organic compounds with low volatility are needed (Kavouras et al., 1998; Zhang et al., 2004; Laaksonen et al., 2008; Hallquist et al., 2009; Jimenez et al., 2009; Riipinen et al., 2012; Ehn et al., 2014; Riccobono et al., 2014; Kirkby et al., 2016; Tröstl et al., 2016; Rose et al., 2018). The most abundant atmospheric oxidants are ozone, OH and NO_3.

The participating compounds differ between the areas. While organic compounds are observed to have a key role in NPF in forestry areas (Laaksonen et al., 2008; Kulmala et al., 2013), some other compounds may dominate the formation and growth in other places, for instance big megacities (Yao et al., 2018), marine atmosphere and clean polar air (O'Dowd et al., 2002; Facchini et al., 2008; Sipilä et al., 2016). For example, iodic acid has found to participate in NPF in coastal areas (Sipilä et al., 2016) and the sulfuric acid-ammonia ion-induced nucleation, followed by sulfuric acid-driven growth dominate NPF in Antarctica (Jokinen et al., 2018).

The role of atmospheric ions has been also under discussion. They are able to stabilize the forming clusters, but also to form neutral clusters by ion-ion recombination (Kazil et al., 2008; Kirkby et al., 2016; Wagner et al., 2017). The ion-induced and ion-mediated nucleation is thought to be still minor compared to neutral pathways (Hirsikko et al., 2011).

Many meteorological, physical and chemical processes, such as presence of clouds (cloudiness vs. clear sky), solar radiation, mixing of air mass, relative humidity and temperature fluctuations, concentrations of participating vapors and condensational sink influence on the occurrence, progress and extent of NPF inside the PBL (Nilsson et al., 2001; Dada et al., 2018). Based on the ground-based and airborne observations it is found that solar radiation, vertical mixing and turbulence enhance NPF being typically higher during NPF event days than on non-event days while relative humidity and condensation sink are lower (Nilsson, 2001; Boy and Kulmala, 2002; Nieminen et al., 2015; Größ et al., 2018). Turbulent mixing may dilute the background aerosols decreasing condensation sink, but in addition it is suggested that turbulence induced fluctuations in temperature and relative humidity can favor the NPF to occur (Nilsson and Kulmala, 1998; Wehner et al., 2010). These conditions could be reached in turbulent layers, such as mixing BL or entrainment zone. The non-event days are observed to be mostly cloudy while clear sky conditions prevail on the NPF event days (Dada et al., 2017).

NPF has been under investigation also in the vertical aspect during past years. Many studies have discussed the occurrence of nucleation mode particles inside the mixing BL (O'Dowd et al., 2009; Crumeyrolle et al., 2010; Mirme et al., 2010; Chen et al., 2018; **Papers I and II**). However, several works have studied aerosol formation in residual layer, RL (Stratmann et al., 2003; Wehner et al., 2007, 2010; Altstädter et al., 2015; Platis et al., 2016), from where the freshly formed particles are able to mix down as the growing BL entrains with the RL. In addition, the freshly formed particles have been observed in outflows of clouds (Clarke et al., 1998; Lee et al., 2004; Wehner et al., 2015) and even FT (Rose et al., 2015; Bianchi et al., 2016; **Paper I**). Accordingly, many studies have been conducted to identify the source region relevant to the atmospheric NPF, and to estimate where the first steps of nucleation takes place. Unfortunately, minimum detection limit of the used instruments has been 3 nm or larger in many of airborne studies, despite the airborne-AIS and NAIS-measurements already in Laakso et al. (2007) and Mirme et al. (2010). In this thesis we succeeded for the first time to measure neutral aerosol particles down to 1.5 nm in diameter from the ground level up to 2700 meters which is crucial in determination of the very first steps of NPF and to identify the regions of interest for the process (**Paper II**).

4.1 Spatial and temporal scale of new particle formation in Hyytiälä

Atmospheric new particle formation (NPF) is frequently observed phenomenon at the ground level at SMEAR II in Hyytiälä. The phenomenon occurs particularly in spring and summer time (Dal Maso et al., 2005; Nieminen et al., 2014; Dada et al., 2017). It was found that meteorological conditions can enhance the formation and growth (Wehner et al., 2010; Platis et al., 2016). Airborne measurements give a possibility to study the regional extent of NPF and especially the measurement of sub-3 nm particles with PSM and NAIS gives important information about the freshly formed particles. With these instruments we are able to get also better estimation about the places that favor new particle formation.

Due to relatively high operation costs, only a limited amount of the airborne measurements are conducted around SMEAR II. Laakso et al. (2007) conducted hot-air balloon measurements around SMEAR II and observed freshly formed charged clusters and neutral nucleation mode particles throughout the whole BL in the afternoon during spring time. The minimum detection limit of neutral particles was 10 nm in diameter with their setup. O'Dowd et al. (2009) reported on vertical profiles during three NPF event days with measurements of >3 nm aerosol particles. Their aim was to find out a possible source region for freshly nucleated particles. They did not observe nucleation mode particles in FT during those days, but detected atmospheric NPF occurring inside the whole ML. However, O'Dowd et al. (2009) found out that in the morning there was a maximum concentration of freshly nucleated particles right above the forest canopy at approximately 30 meters above ground. They speculated that the possible source for precursor gases could be the forest and the particles and precursors were subsequently mixed up within the rising BL. A quite large study of nucleation mode particles over SMEAR II is also presented in Väänänen et al. (2016) supporting the hypothesis that the nucleation mode particles are formed mainly inside the BL, but sometimes they are observed also separately in RL and even FT.

In this thesis we present the observations from the measurement campaigns in 2009, 2010, 2015 and 2017 (**Papers I, II and IV**). The analysis in **Papers I** and **II** target different NPF event days. While the studied measurement flights in **Paper I** were conducted in June, September and October, the flights in **Paper II** were conducted mainly in spring and summertime. Instrumentation was also different between the campaigns. Prior to this thesis, the vertical variability in the concentration of neutral sub-3 nm particles have not been under investigation around the SMEAR II. Starting from

2015 the neutral particles down to 1.5 nm in diameter were measured with the Particle Size Magnifier (PSM).

During typical day in spring-summer time the daytime BL evolution starts in the morning after sunrise in Hyytiälä. The particle population gets mixed inside the evolving BL and BL height can extend up to 1000–2000 meters during the day (see Fig. 4 in **Paper II**). When thermal convection and turbulence mix the BL air, vapors, moisture and aerosol particles mix up from the ground level, but there is also concurrent mixing down from the RL and FT region in entrainment zone. A typical NPF event day in Hyytiälä is sunny and non-cloudy spring or summer day (Nieminen et al., 2015; Dada et al., 2017), when air masses arrive from relatively clean sector, originating from North Atlantic Ocean or arctic areas being transported over boreal forest for several hours (Sogacheva et al., 2005; Dal Maso et al., 2007; Nieminen et al., 2015; Dada et al., 2017; **Paper I**).

Because of many of the earlier observations have been case studies, we combined in **Paper II** several vertical measurement profiles to study the median vertical and temporal behavior of the particle concentrations. We found that the total particle concentration was significantly higher inside the BL than above it. The smallest nucleation mode particles occurred mainly throughout the whole BL and the highest concentrations were observed at lower altitudes. The results are in line with other airborne observations about the NPF around SMEAR II (Laakso et al., 2007; O'Dowd et al., 2009; Väänänen et al., 2016). The maximum in the median concentration of 1.5–3 nm particles is consistent with the idea that turbulent mixing distributes vegetation originated NPF precursor gases from the canopy upwards within the mixing BL and possible the formation starts in the turbulent layer above the forest canopy top. Same kind of vertical distribution of sub-3 nm particles was seen in a case study presented in **Paper II**. However, other pathways for the NPF events are also possible, such as RL originated gaseous compounds mixing down into the turbulent ML where they are able to initiate formation. NPF may also start already in RL or turbulent entrainment zone and freshly-nucleated particles and gaseous precursors can mix down to the BL where they can continue to grow if condensing vapors are available (see e.g. Nilsson et al., 2001). Prevailing formation processes and places, where formation and subsequent multicomponent condensational growth can take place depend on e.g. the ambient vapor concentrations, background aerosol concentration and meteorological conditions.

In our study, we defined the median vertical profile for the particle number concentration separately in the size ranges of 1.5–3 nm and 3–10 nm (**Paper II,** Fig. 2 and 3 there). Our aim was to investigate the possible sources for the gaseous precursors for NPF and initial places where nucleation takes place and how NPF proceeds in the boreal forest environment. We tried to find possible similarities and differences between morning and afternoon times, separately for the NPF event and for the

undefined days. The event classification was done based on ground-based observations. The event classification is discussed in more detail in Chapter 4.2. In short, we divided the total 53 measurement flights as morning and afternoon flight profiles. The data used in Fig. 2 and 3 in **Paper II** is collected to the Table 4.1 (here). The vertical behavior of the concentration of sub-3 nm particles was similar in the mornings and afternoons of the NPF event days and undefined days. The concentration of sub-3 nm particles was, on average, were higher at lower altitudes decreasing with increasing altitude. This is consistent with the hypothesis that the oxidation products of volatile organic compounds originating from the vegetation could contribute significantly to the NPF process over boreal forest, already suggested in early studies based on the ground level measurements (Tunved et al., 2006; Paasonen et al., 2010).

During the NPF afternoon times, less vertical difference was present in the median of sub-3 nm particle concentration inside the BL, which is explained by the more effective mixing within the BL (see also Table 4.1). Comparing the morning time median profiles between the NPF event and undefined days, we can see that the maximum in the vertical profile of 1.5–3 nm particle is right above the forest canopy top in the NPF event days whereas a corresponding small decrease is seen in the undefined day morning profile (Fig. 3 in **Paper II**). This is possibly explained with a different kind of formation processes for nucleation mode particles between NPF event and undefined days.

Table 4.1. Statistics about the study used also in Fig. 2 in **Paper II**.

	Number of flight profiles	Median conc. (1.5–3 n) inside BL (cm^{-3})	Median BLH (m)	Median SHF (Wm^{-2})
All days	**53**	**2260**	**1400**	**174.6**
morning	27	2754	1100	162.7
afternoon	26	1404	2000	200
NPF events	**17**	**1743**	**1300**	**200**
morning	9	1743	800	165.8
afternoon	8	2346	1775	286.2
Undefined days	**34**	**2384**	**1400**	**167.7**
morning	18	2866	1100	159.5
afternoon	16	1404	1600	172.3

We found relatively higher concentration of sub-3 nm particles during the NPF event afternoons and during undefined day morning times in the ML above the canopy. As we see in Fig. 2 and 3 in **Paper II**, freshly formed particles were observed in the ML during several undefined days but the growth was typically interrupted, possible due to the lack of gaseous precursors or changes in favorable meteorological conditions for the condensation (see Buenrostro Mazon et al., 2009; Kulmala et al., 2013). The aerosol particle formation and growth, in turn, start often later during the NPF event days than undefined days, approximately around noon at SMEAR II.

We compared the median concentration of sub-3 nm particles on the ground and inside BL over SMEAR II (Table 1 in **Paper II**). The dataset is limited to the cases of available on-ground data. We found out that, on average, the concentration of sub-3 nm particles was lower on the ground level than it was inside the BL above the canopy. A similar observations was reported in Väänänen et al. (2016). The results can be explained by increased sinks for the particles and precursor vapors inside the canopy. In addition, it has to be recognized that also the different instrumentations on the ground and on the plane introduce uncertainties in the comparison. From the study we can summarize that that the concentration of sub-3 nm particles was on average higher above the canopy in the ML during undefined day morning time in comparison with the concurrent data at the ground level. This points towards a suitable place for the nucleation above the canopy, not inside it, which was hypothesized already in Nilsson et al. (2001).

The case studies in **Papers I and II** underline the existence of new nucleation mode particles in RL or FT, separately from the NPF inside the mixing BL (Fig. 2 and 3 in **Paper II**). In the case study in **Paper II** the presence of sub-10 nm particles were observed in 2500 meters while the new BL evolved below 700 meters and formation of sub-3 nm particles occurred inside the ML. In the case studies in **Paper I**, the nucleation mode particles were observed in 2500–3500 meters, separately from the ML particle formation. If these particles are not removed by deposition or coagulation before the new BL reaches the height of these layers, the nucleation mode particles may mix down and continue to grow in size in the ML (transported events, see more Dada et al., 2018).

Although the nucleation mode particles were observed in the ML, RL and even FT, the progress of the processes and thus place where the formation and growth takes place and where the participating gaseous precursors are from have still remained a partly open question. One explanation is that the nucleation and growth starts in RL/turbulent inversion layer, where the gaseous precursors and particles from the previous' day BL remain throughout the night. There they can undergo further oxidation producing more precursor vapors and start to nucleate and grow later on, enhanced with the onset of photochemistry in the morning. In the mixing air masses, turbulence can generate

supersaturated regions favoring sporadic nucleation. The freshly formed particles can entrain to the BL where for instance the oxidized organic precursor gases condense onto the particles and they grow in size (Wehner et al., 2010). Other suggestion is that vegetation originated vapors emitted by the biosphere mix upwards in the morning and start to nucleate in the turbulent BL. The air above the forest canopy favors NPF compared to the forest canopy because the photochemistry is more efficient above the canopy than inside the canopy (Hens et al., 2014) and the canopy sink for the particles and precursors is reduced (Rinne et al., 2012). Furthermore, there is a lower background aerosol concentration and consequent condensation sink for the vapors above the canopy than inside the canopy.

However, in order to better define and recognize the different nucleation and growth mechanisms and participating precursor gases in the Hyytiälä area and clarify the places where nucleation starts (e.g. mesoscale vortices, Lampilahti, MSc. Thesis, 2016), we would have to conduct airborne measurement flights with instruments that can identify the gaseous compounds together with aerosol and meteorological instrumentation.

4.2 Identifying new particle formation events from intermediate ion concentration

The occurrence of an NPF event has traditionally been identified based on a visual estimation from ion and total number size distributions. The method is well-described in Dal Maso et al. (2005) and Hirsikko et al. (2007). The criteria for an NPF event day are i) detection of a new mode of particles in nucleation mode size range, ii) the mode lasts for 1 hour or more and iii) the growth of those particles is observed during the following hours. According to the traditional NPF event classification method the days are divided into NPF events, non-events and undefined days. This method was used in in **Papers I and II**.

It is recognized that visual and subjective way in traditional classification gives some uncertainties. In addition, the method is very slow to use for long time series. In this thesis we developed a new statistical and automatic identification method for the NPF events (**Paper III**). It is based on an increase in intermediate size range ion concentration when the NPF event is ongoing. The size range of intermediate ions is ~2–7 nm and they form a narrow band of ambient naturally charged particles right above the cluster ion band. The method relies on the fact that the concentration of intermediate ions is typically very low (< 10 cm^{-3}, Tammet et al., 2014). The concentration of intermediation ions

is higher during the NPF events (Hirsikko et al., 2011) or during rain (Tammet et al., 2009; Kolarž et al., 2012).

Fig. 4.1 depicts a time series of number concentrations of negative and positive intermediate ions at SMEAR II. This is the example of the occurrence of daytime and night time NPF. The ions are suggested to be in a more significant role in the nocturnal than daytime nucleation process at SMEAR II (Junninen et al., 2008; Rose et al., 2018), but the classification method is valid also for the daytime NPF events. The nocturnal cluster events start usually in the evening, and the formed clusters rarely grow past a few nanometers (Buenrostro Mazon et al., 2016) in size.

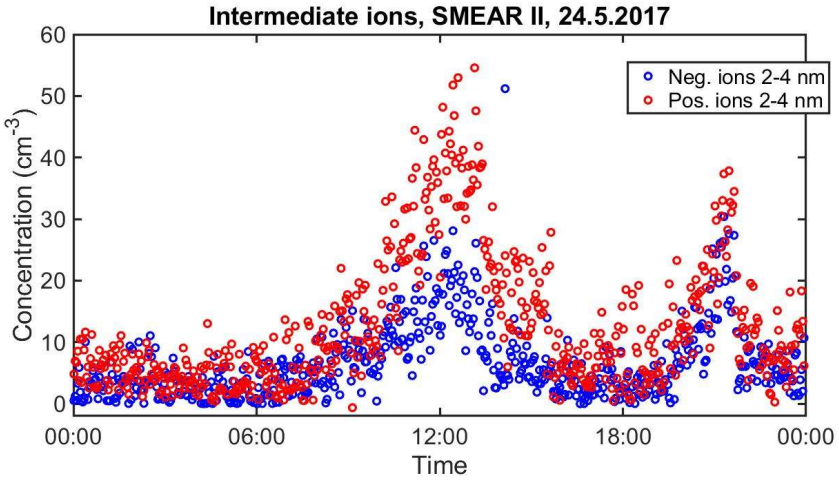

Figure 4.1. Time series of intermediate size range positive and negative ions measured at SMEAR II in Hyytiälä on 24.5.2017.

Both polarities of NAIS ion data are suitable for the NPF event identification. The Earth surface is negatively charged and therefore attracts positively charged particles. Rain is been identified as a source for the negatively charged particles, also in intermediate ion size range (Tammet et al., 2009; Kolarž et al., 2012). This needs to be taken into account by utilizing meteorological observations to support the NPF identification with this method. With this new method, we are able to get information about NPF event and non-event times analyzing the measured concentrations of intermediate ions, ignoring the precipitation times.

We tested the new method for a long time-series of naturally charged ambient intermediate ion concentrations measured at SMEAR II in Hyytiälä between the years 2003–2013 in **Paper III**. The data was measured with Neutral Cluster and Air Ion Spectrometer (NAIS, Manninen et al., 2016). The instrument is described in Section 3.3.1. We calculated 30-minute and 1-hour median

concentrations of the negative intermediate ions and examined how well the results agree with traditional classification. We varied the threshold concentrations for the intermediate ions to classify the NPF occurrence. We found 77.5 % agreement between the methods with a concentration of 20 cm^{-3} as the threshold (Fig. 4.2a). If we use the size range of 2–7 nm instead of 2–4 nm in the criteria, the agreement with the traditional method is even better, 92.3 %. The new method is compatible with the criteria in traditional method about the observed growth of newly formed particles during NPF event day.

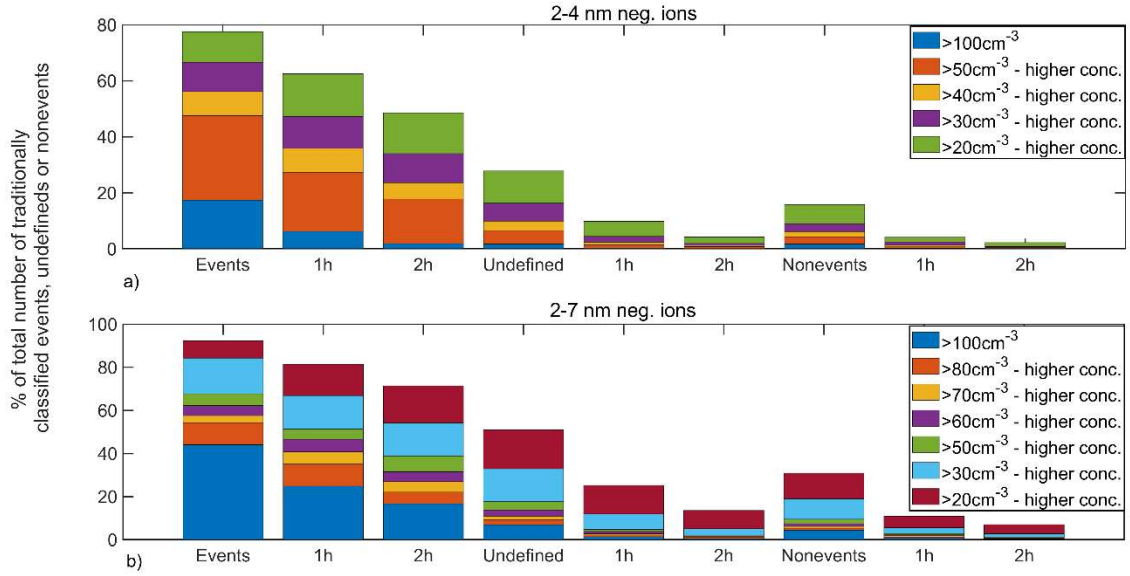

Figure 4.2. Bar charts in this figure represent the similarities in two NPF event classification methods. In Fig. a) we compared used the concentrations of 2–4 nm negative ions measured at daytime between 6:00–18:00 with variable threshold concentrations. Fig. b) is the same but with the 2–7 nm negative ion concentrations. The dataset includes in total 702 NPF events, 1090 undefined days and 976 non-event days during the years 2003–2013 at SMEAR II classified with the traditional method. The bars "Events", "Undefined" and Nonevents" represent the half hourly medians used in this study followed by bars representing longer averaging times (1h and 2h) for these cases.

We compared the results of the two NPF event classification methods as a function of season. The analysis revealed that that both methods give the highest number of NPF events during spring time and the variability over the year was similar, which supports the reliability and usefulness of the new NPF classification method. The best agreement with the traditional method was with using 30 cm^{-3} as the limit concentration of 2–4 nm negative ions or correspondingly threshold of 50–60 cm^{-3} in case

of the 2–7 nm negative ions. In over 40 % of the traditionally classified NPF event days the half hourly median concentration of 2–7 nm negative ion exceeded 100 cm^{-3} during the daytime event (Fig. 4.2b).

We investigated the diurnal variability of half hourly median concentration of 2–4 nm negative ions measured at SMEAR II during 2003–2013, separately for NPF events, undefined days and nonevent days classified by traditional NPF event classification method (Fig. 4.3). The concentration of 2–4 nm ions had a diurnal maximum at the same time, when the NPF event is typically detected at SMEAR II. The concentration started to increase around 7 o'clock in the morning local time. A modest increase was also apparent in the evening, which indicated nocturnal nucleation (Junninen et al., 2008). The interesting diurnal behavior was detected during the undefined days, particularly in the 75th-percentiles, which increased in the evening. This indicated nocturnal formation also during undefined days. With the traditional method, the nocturnal NPF events are not classified as NPF events as they typically do not show growth of the clusters (Mazon et al., 2009).

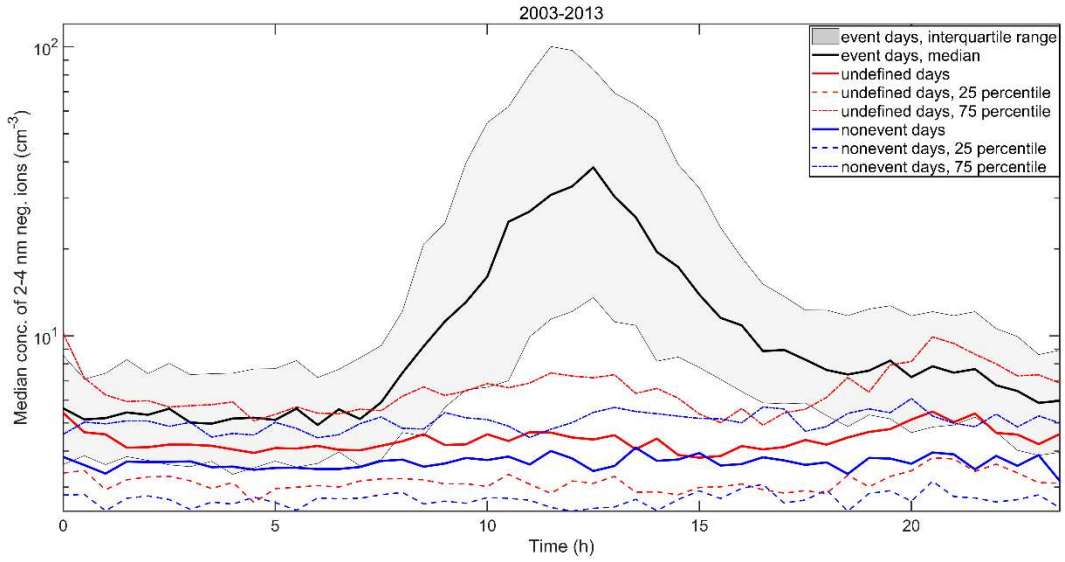

Figure 4.3. The median diurnal cycle of 2–4 nm negative ion concentration during NPF events, undefined days and non-event days and their interquartile ranges (25th and 75th percentiles).

As a summary, the new method is complementary to the traditional NPF event analysis as the benefit is a quick, automatic way to process large data sets. Disadvantages compared to the traditional method is that the method relies on the availability of the ion size distribution data and supporting data on precipitation.

In addition, the new method differentiates the NPF events and the non-events depending only based on the appearance of intermediate ions. Therefore, implicitly it does not detect or in any way consider the growth of particles, which is one of the criteria in traditional NPF event classification. Furthermore, sometimes it is seen that the NPF starts from larger sizes, but the growth is observed similar to a typical NPF event. Such "tail events" may possibly be induced and enhanced by differences in horizontal advection (change of air mass) or vertical mixing, which brings gases and particles from upper parts of atmosphere. The method in **Paper III** is unable to detect this phenomenon as it is associated with lack of increased intermediate ion concentration and consequently the method classifies them as non-events. As a next step and building upon **Paper III**, a refined new particle formation classification scheme was developed by Dada et al., 2018.

5 Long-range transported air pollution from biomass burning

Biomass burning produce particulate matter in large quantity and emit also gases like carbon monoxide (CO), carbon dioxide (CO_2), water vapor (H_2O), ozone (O_3), nitrogen and sulfur-containing species into the atmosphere (Andreae and Merlet 2001; Langenfelds et al., 2002; Jaffe et al,, 2004; Niemi et al., 2005; Reid et al., 2005; van de Werf et al., 2010; Portin et al., 2012; Krol et al., 2013; **Paper IV**). The burning produces carbon and nitrogen species, volatile organic compounds (VOC) and particulate matter (Lemieux et al., 2004). The aerosol particles from biomass burning are typically in Aitken and accumulation mode size range, but also nucleation mode particles have been observed near active fire areas (Virkkula et al., 2014). Overall, biomass burning produces both primary and secondary aerosol particles (Heringa et al., 2011).

The aerosol particles and trace gases can travel long distances from their source regions (Salvador et al., 2013; Ancellet et al., 2016; Chouza et al., 2016). When intensive wild fires occurred in Eastern Europe in spring 2006 and late of summers of 2006 and 2010, the aerosols and trace gases emitted by the fires were detected hundreds or even thousands of kilometers away from fire areas (Saarikoski et al., 2007; Stohl et al., 2007; Witham and Manning, 2007; Anttila et al., 2008; Portin et al., 2012; Mielonen et al., 2012, 2013; **Paper IV**). Transport of the pollutants depends on the meteorology, like air mass movement, but also lifetime and reactions of pollutants in the atmosphere.

As part of this thesis, we investigated long-range transported anthropogenic air pollutants released by intensive wildfires in Eastern Europe. Associated to the fires, we detected elevated concentrations at three SMEAR stations and on-board Cessna during flight measurement campaign in Finland (**Paper IV**). We found that long-range transported smoke plumes included elevated concentrations of CO, CO_2, SO_2, O_3, NO_x, aerosol particle number concentrations and black carbon mass concentration at ground stations in different part of Finland (Table 5.1 and other tables in **Paper IV**). We compared the aerosol and trace gas data during the long-range transport episodes with typical concentrations for each season and site. As the reference period for the selected quantities on smoke days we used averages of July-August period over 5–14 years, depending on availability of data.

We observed that the aerosol particle median size was 60–210 % larger in Hyytiälä during the studied smoke days than it was during the corresponding reference period (Fig. 5.1 and Table 5.1). The median particle size peaked in accumulation mode size range. The total number concentrations measured at all stations were also elevated compared to the corresponding concentrations during the reference period. The analysis of air mass origin with back trajectories (Draxler and Hess, 1998; Stein

et al., 2015; Rolph, 2016) supports the observations that the detected particles and gases came from south or southeast where the wildfires occurred.

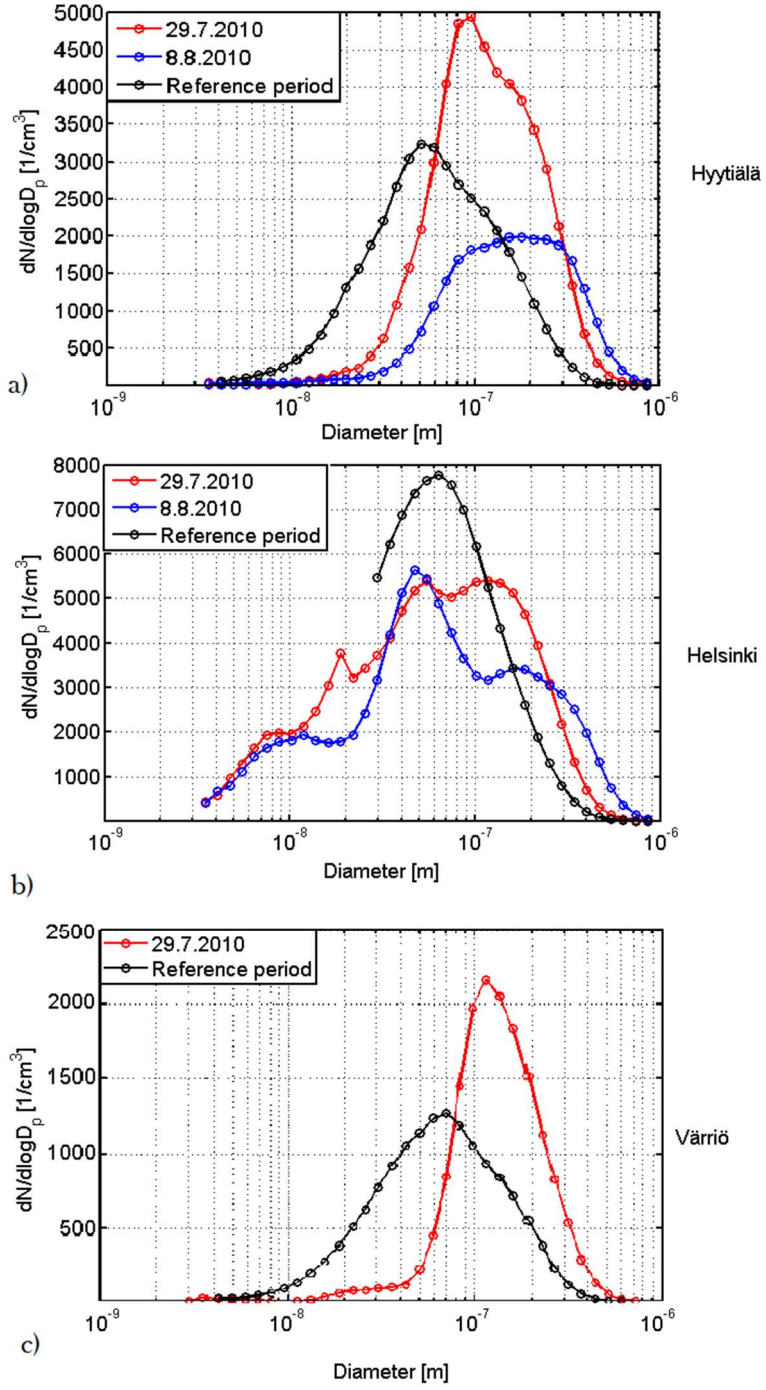

Figure 5.1. Median size distributions in all three SMEAR stations in Finland during two the most intensive smoke days in 2010 and over reference period (modified version of the figure in **Paper IV**).

Based on aerosol optical measurements at Hyytiälä, the smoke was observed to be highly processed. The values of single scattering albedos were high during both the most intensive smoke days selected for a closer review in this study. Together with high concentration of BC and high single scattering albedo are indicative of strong secondary aerosol formation in the transported fire plumes (Liu et al., 2013; Vakkari et al., 2018). In addition, we found a high Ångström exponent of scattering, which is explained by the domination of small particles, concurrent with a high Ångström exponent of absorbing indicating a high fraction of organics in the particles (**Paper IV**).

However, it should be noted that during the transport time from the fire areas, the air mass is affected by other emission sources along the way, such as industry and traffic. They produce e.g. NO_x, SO_2 and aerosol particles, which contribute to the net effect observed at the receiver site.

Table 5.1. Mean values of different trace gas and total particle concentrations, median aerosol particle size and mean condensation sink observed at SMEAR II in Hyytiälä during the intensive smoke days in 2006 and 2010 compared to the reference values (adapted from **Paper IV**).

Hyytiälä	2 May 2006	14 Aug 2006	29 July 2010	8 Aug. 2010	Ref.	Ref. period
CO (ppb)	259	298	262	276	118	2002–2010
CO_2 (ppm)	381	371	396	405	372	1996–2010
SO_2 (ppb)	1.26	0.21	1.34	0.35	0.13	1997–2010
O_3 (ppb)	65.3	34.0	55.7	42.0	30.8	1996–2010
NO (ppb)	0.01	0.01	0.02	0.02	0.02	1996–2010
NO_x (ppb)	3.05	1.82	0.41	0.55	0.95	1996–2010
H_2O (ppth)	4.7	13.3	16.5	18.8	12.5	1996–2010
BC (ng m^{-3})	1626	839	1054	1270	240	2005–2010
D_p (nm)	129	146	117	152	73	1996–2010
N_{tot} (cm^{-3})	5558	2983	3496	1806	2139	1996–2010
CS (s^{-1})	0.019	0.014	0.012	0.011	0.004	1996–2010

In addition to the ground-based observations, four aircraft measurement flights were conducted during the intensive fire period in 2010 in Eastern and Central Finland. We detected clear smoke plumes over Jämsä area on 29th July 2010. During this time, total particle concentration exceeded 6000 cm^{-3} at 2500–3500 meters above ground level in the afternoon and the smoke was detected also by a distinct odor by the operator and the pilot. During that flight, the total number concentration was higher than typical concentration (100–2000 cm^{-3}) in FT (**Papers I, II and IV**). We compared the

aerosol number concentration aloft in the smoke plume to the particle number concentration at SMEAR II, Hyytiälä and noticed that the aerosol and smoke was more prominently present at higher altitudes than at the ground level. Based on satellite derived aerosol optical depth with MODIS (Moderate Resolution Imaging Spectroradiometer), the smoke plume was not regionally homogenous in Southern Finland. In addition to the horizontal variability, there were also vertical differences. If the convective mixing is weaker or the BLH does not reach the layers aloft, the biomass burning aerosols can travel over the area undetected at the ground level. The distance between observations in Jämsä and Hyytiälä is ~50 km.

As a summary from **Paper IV**, we concluded that smoke plumes from intensive wild fires detected in Eastern Finland were transported hundreds or even thousands of kilometers from the source areas. Comparing the size distributions at three measurement stations in Finland we found that the median particle size grew in diameter during the transport. The aerosol number concentration peaked in accumulation mode size range and the total particle concentrations were higher than on average at all the stations. Based on the aerosol optical properties we found the smoke plumes were strongly scattering and secondary aerosol production and condensational growth with organic compounds were probable. Although the total effect on climate was probably cooling, the absorbing BC was produced by the fires as well.

6 Review of papers and the author's contribution

Paper I investigates the spatial and temporal variation of nucleation mode particles at SMEAR II. The analyzed data was collected using the Cessna aircraft as a platform. I participated in the measurement campaign and contributed to the analysis and writing of the manuscript.

Paper II studies vertical extent of newly formed clusters, median total particle concentrations in different altitudes along the day above the SMEAR II, Hyytiälä. One case study of a developing BL associated with aerosol particle mixing in the vertical during new particle formation event was examined in more detail. I was responsible for the preparations and operation of the experimental aircraft operation and campaigns together with Riikka Väänänen and Janne Lampilahti. I contributed to the development of the instrumentation on board the Cessna. I conducted the data analysis process and wrote most of the paper.

Paper III studies concentrations of intermediate ions related to the identification of new particle formation events. The data was measured with Neutral Cluster and Air Ion Spectrometer (NAIS) at SMEAR II in Hyytiälä over several years. The concentration of intermediate ions in size range of 2–4 nm was observed to be a strong indicator of NPF bursts. I conducted the whole data analysis and wrote the paper.

Paper IV studies long-range transported biomass burning pollutants in Finland during intensive forest fires in Eastern Europe in the years of 2006 and 2010. I analyzed the gas and particle data sets measured at SMEAR stations and Cessna and wrote the initial manuscript. Laura Riuttanen was responsible for the satellite data analysis and backward trajectory analysis in paper and wrote the final version of the paper.

7 Conclusions

In this thesis I studied both new particle formation events (NPF) in spatial and temporal scale in relatively clean boreal forest environment and long-range transported air pollution to Finland originating from intensive wildland fires in Eastern Europe. The observations were done at SMEAR stations in Finland and onboard Cessna research aircraft. The NPF was studied around SMEAR II in Hyytiälä.

We found that the air masses originating from fire areas far away showed elevated concentrations of different trace gases and aerosol particles including black carbon. We found the particles grew in size during the transport and the size distribution was dominated by the accumulation mode sized particles. The particles were observed to have a high single scattering albedo contributing to a cooling effect although the co-emitted black carbon, in contrast, has a warming effect via absorbing the solar radiation. We found that the particulate matter originating from Russian forest fires was, on average, more scattering than absorbing after they travelled to Finland. We observed that the transported aerosol particles were not divided homogenously over Southern Finland, but occurred in plumes instead. One particularly intensive smoke plume with high concentration of aerosol particles was detected during Cessna measurement flight at 2500–3500 meters a.g.l over Jämsä area.

Our findings provide experimental data on dispersion of pollution from wide uncontrolled biomass fires that can affect both people's health and the environment even hundreds of kilometers away from the fire areas. As the climate change increases risk for droughts (Naumann et al., 2018), it is plausible that more fire plumes reach Finland in the future. Although it is general knowledge that fires produce climate-warming greenhouse gases, they also produce a lot of particles that are able to accelerate or slow down the climate change. The net effect needs to be refined when more scientific data is available.

In this thesis, the airborne and ground-based measurements are used also in the basis of studying of another source of atmospheric aerosols, new particle formation. We studied NPF events over SMEAR II area during several airborne measurement campaigns between 2009 and 2017. Many studies have shown that suitable conditions and contributors for enhancing the formation and growth of new atmospheric aerosols can occur in different places within the boundary layer: in the BL, RL, in the entrainment zone and even in the FT. Observations of NPF in mixed layer have many reasons. For example, when thermal convective mixing dilutes the aerosol population inside the ML, the conditions for nucleation and growth can become favorable due to lower condensation sink. Secondly, turbulent mixing is likely to lead to higher localized super saturation pockets of possible gaseous

precursors which are essential for NPF. If the layer is well mixed, observed NPF event can extend to occur throughout the whole BL.

The first step in the investigation of NPF is to analyze, if the day is NPF event or non-event or something between those two categories. To facilitate and automate the event day classification, we developed a new objective and automatic classification method which utilizes the measured ion concentration in the intermediate size range, 2–7 nm as an indicator for the NPF periods. The concentration in this size range is generally low, if NPF does not occur. We found that the intermediate ion concentration was a suitable parameter to recognize the NPF events and to separate them from the undefined days and from the non-event days. Recently, the objective classification and characterization of the NPF events was further improved by a refined, automated method that recognizes also transported NPF events and ion bursts and separates them from the regional NPF events (Dada et al., 2018). This new method characterizes and classifies also the undefined days, which is a substantial improvement. Both methods are based on the air ion concentrations in intermediate ion size range as the suitable indicator for NPF.

Our findings in this thesis support the concept of NPF occurring within the BL in quite clean boreal environment, where the background aerosol concentration is comparatively low. We found that during NPF event days in Hyytiälä, the particle formation frequently starts right above the forest canopy top and extends to cover the whole BL, when the convection mixes the air inside the developing BL. The organic condensable vapors originating from oxidation products of biospheric emissions probably have an essential role in these regional NPF events. However, we suggest that NPF does not necessarily start every time in the lower part of the BL. We found signs that sometimes the formation of the smallest particles could initiate at a higher altitude and the particles then mix down to the BL where they can continue to grow, if the precursors are available and thermodynamic conditions favor multicomponent condensation. This kind of events can be classified as transported NPF events (either vertically or horizontally, or both). The likely places for the nucleation in these cases are Residual Layer (RL), Entrainment Zone (EZ) or Free Troposphere (FT). The contributors for this kind of formation could be the gaseous precursors that originate from the RL that developed during the previous day.

Our results improve our understanding of the first steps of atmospheric NPF inside the BL and shed light on the connection between boundary layer dynamics and NPF. Next step is to investigate different formation pathways in more detail. To achieve this, it would be important to study the vertical variation of particle concentrations in different size ranges and their chemical composition with high resolution mass spectrometry, which would enable us to assess more specifically the

possible sources of the aerosol particles and their precursor gases. In addition, the contribution of mesoscale convection induced movement, like roll vortices to the NPF needs to be investigated in more detail (Lampilahti, MSc. Thesis, 2016).

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